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A STUDY OF THE MECHANISM OF EXPLOSIVE COMBUSTION OF GAS MIXTURES

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/Figures are appended./

/This paper clarifies the mechanism of explosive combustion of gas mixtures, refers to the role which nitrogen plays in the process under consideration, and explains the inhibiting action of tetraethyl lead in explosive gas mixtures on the basis of new experimental data/.

In several investigations of the mechanism of nitrogen oxides formation (2-3) during the explosive combustion of methane in the presence of both oxygen and nitrogen, we obtained results which induced us to formulate the question in regard to the transformation of the chain mechanism into a thermal mechanism with increased pressure and increased dimensions of reaction vessels.

It was established that with increased pressures and dimensions of vessels, the yield of nitrogen oxide first rises to values of the order of 1.5-2.0% and then drops to 0.4-0.5%, i.e., to yields which are characteristic of pressures of the order of one atmosphere and large reactors. This dependence of the yields of nitrogen oxide is difficult to explain on the basis of either the chain theory of explosions (4) or the thermal theory (5). The observed dependence, in our opinion, must necessarily arise in connection with transformation of a chain mechanism into a thermal mechanism taking place under the effect of triple collisions, this transformation being favored by increased pressures and larger vessels.

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Nitrogen participates in these triple collisions, dissociating into atoms and being then transformed into nitrogen oxide, so that with increased pressures and vessel dimensions the yields of this compound are increased. The yields rise to a maximum representing an upper limit and then begin to drop.

The drop of the yield coincides with beginning preponderance of the thermal mechanism, lower yields of nitrogen oxide being characteristic for the latter.

D. F. Frank-Kamenetskiy (6) holds that the dependence of the yield of nitrogen oxide on the diameter of the vessel is characteristic only for combustible mixtures of intermediate composition. According to Frank-Kamenetskiy, mixtures of intermediate composition behave in small vessels as if they were poor in combustible substances (under such conditions the yield of NO increases with the diameter of the vessel), while in large vessels they behave as mixtures which are rich in combustible substances, i.e., the yield of NO passes through a medium and then drops. It seems to us that the assumption of an influence of the diameter of the vessel on the type of composition of a combustible mixture is hardly satisfactory, especially since it has not been proved. On the other hand, we have established the occurrence of an optimum yield depending on the diameter with combustible mixtures having any composition.

Some of the results proving our point are cited below. Our method of investigation has already been described (1) and another description of the method will be given in a future paper presenting all the results obtained hitherto.

Figure 1 shows the dependence of the yield of nitrogen oxide on the concentration of admixed tetraethyl lead in the explosive combustion of hydrogen. The composition of the mixture tested was 38% H_2, 40% O_2, and 22% N_2. It can be seen from Figure 1 that the yield of nitrogen oxide depends very strongly on the concentration of tetraethyl lead. This dependence is most pronounced at low pressures, when the chain mechanism is more strongly developed. Tetraethyl lead apparently participates as an active inhibitor in the triple collisions involving N_2 which lead to the formation of NO. Under the circumstances, the probability that NO will form is lowered.

Figure 2 shows the dependence of the lower limit of spark ignition on the concentration of tetraethyl lead in a gas mixture of the same composition. It can be seen that this dependence is rather pronounced. The lower limit is raised by 20-30 mm with an increase of the concentration of tetraethyl lead from 0.0034 to 0.034%. Tetraethyl lead obviously has the effect of an active inhibitor which interrupts chains and makes the ignition of a combustible mixture more difficult. Tetraethyl lead already exerts its maximum effect at low concentrations, just as do other inhibitors. Figures 1 and 2 show that the reactions of explosive combustion have a chain character at low pressures.

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/Figures follow./

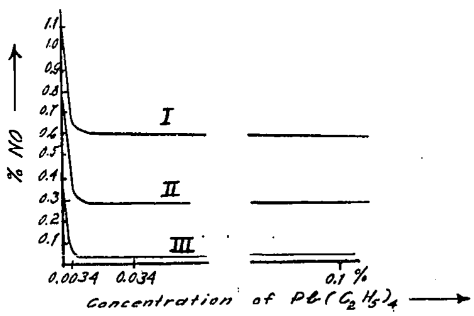

Figure 1. Dependence of the Yield of Nitrogen Oxide on the Concentration of Tetraethyl Lead at the Following Pressures: I - 300 mm Hg; II - 200 mm Hg; III - 100 mm Hg.

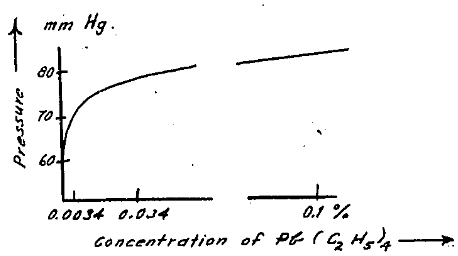

Figure 2. Dependence of the Lower Limit of Spark Ignition of the Mixture 38% H_2, 40% O_2, 22% N_2 on the Concentration of Tetraethyl Lead.

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